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16. ABSTRACT

This report discusses environmental levels of methylchloroform (MC) based on a review of the literature and other information sources. The concentrations of MC in the U.S. atmosphere ranges from about 0.1 $\mu g/m^3$ (20 ppt) in remote areas to over 500 $\mu g/m^3$ (100 ppb) in some areas near where the substance is manufactured or used. The concentration drops off rapidly as one moves away from a source facility. Surface water contamination of MC range from somewhat less than 1 ppb to several hundred ppb in the vicinity of MC manufacturers. The highest measurement reported (3 ppm) was made in a roadside ditch near a producer site. MC has been detected but not quantified in U.S. drinking water except in one case when approximately 10 ppb was reported. Soil and sediment concentrations of MC appear to be no higher near manufacturers and users than in rural areas, though the data are very limited. The levels are on the order of fractions of a ppb. MC is a saturated chlorinated hydrocarbon which is relatively stable in the atmosphere. However, the molecule is susceptible to hydrolysis or dehydrohalogenation and reacts with water relatively rapidly and is thus degraded in soil and water. There are very few data on presence of MC in food raised and sold in the U.S. However, data from the United Kingdom suggest that MC is found on the order of parts per billion in some common foodstuffs. There is little evidence to judge whether MC accumulates in living organisms. Limited data on levels in marine organisms show levels on the order of a few parts per billion.

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MULTIMEDIA LEVELS
METHYLCHLOROFORM

September 1977

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EXECUTIVE SUMMARY

This report discusses environmental levels of methylchloroform, based on a review of the literature and other information sources.

The concentrations of methylchloroform in the atmosphere of the U.S. range from about 0.1 $\mu g/m^3$ (20 ppt) in remote areas to over 500 $\mu g/m^3$ (100 ppb) in some areas near where the substance is manufactured or used. The concentration drops off rapidly as one moves away from a source facility.

Surface water concentrations of methylchloroform range from somewhat less than 1 ppb to several hundred ppb in the vicinity of methylchloroform manufacturers. The highest measurement reported (3 ppm) was made in a roadside ditch near a producer site.

Methylchloroform has been detected but not quantified in U.S. drinking water except in one case when approximately 10 ppb was reported.

Soil and sediment concentrations of methylchloroform appear to be no higher near manufacturers and users than in rural areas, though the data are very limited. The levels are on the order of fractions of a ppb.

Methylchloroform is a saturated chlorinated hydrocarbon which is relatively stable in the atmosphere. However, the molecule is susceptible to hydrolysis or dehydrohalogenation and reacts with water relatively rapidly and is thus degraded in soil and water.

There are very few data on the presence of methylchloroform in food raised and sold in the U.S. However, data from the United Kingdom suggest that methylchloroform is found on the order of parts per billion in some common foodstuffs.

There is little evidence to judge whether methylchloroform accumulates in living organisms. Limited data on levels in marine organisms show levels on the order of a few parts per billion.

1. INTRODUCTION

Methylchloroform (MC) is one of the chemicals whose health and ecological effects, environmental behavior, and technologic and economic aspects are important to the U.S. Environmental Protection Agency. The literature has been searched in an effort to determine the environmental levels of methylchloroform, the behavior of methylchloroform in the environment, and the ways in which methylchloroform may come in contact with man.

The literature has been examined using the following search strategy. An initial computer search of the following data bases was conducted:

- National Technical Information Service (NTIS)
- Smithsonian Science Information Exchange (SCD-SSIE)
- Engineering Index
- Pollution Abstracts
- TOXLINE
- MEDLARS (National Library of Medicine's National Interactive Retrieval Service)
- Air Pollution Technical Information Center (APTIC)
- USGS Water Quality Monitoring Data.

All searches were carried out in June, 1976. Original journal articles with relevant titles or abstracts were then examined and data extracted. In addition, various journals were screened manually through December, 1976. These journals included: Analytical Chemistry, Atmospheric Environment, Bulletin of Environmental Contamination and Toxicology, CRC Critical Reviews in Environmental Control, Environment, Environmental Pollution, Environmental Research, Environmental Science and Technology, International Journal of Environmental Analytical Chemistry, Journal of Environmental Science and Health, Journal Water Pollution Control Federation, and Water Research. Other journals were also screened but are not listed because they did not cover the indicated period, or were of more limited interest to those seeking information on environmental levels of methylchloroform.

Several important reviews on the subject of methylchloroform were also consulted. Specifically, a preliminary study of selected potential

environmental contaminants including methylchloroform (U.S. Environmental Protection Agency, 1975a), a preliminary economic impact assessment of possible regulatory action to control atmospheric emissions and selected halocarbons (Shamel, 1975), a criteria for a recommended standard for occupational exposure to 1,1,1-trichloroethane (methylchloroform) (U.S. National Institute for Occupational Safety and Health, 1976), and a toxicology study called "Methylchloroform and Trichloroethylene in the Environment" (Aviado et al., 1976) were consulted.

2. OCCURRENCE OF METHYLCHLOROFORM IN THE ENVIRONMENT

METHYLCHLOROFORM IN THE ATMOSPHERE

No extensive monitoring program designed specifically for methylchloroform has been identified. However, methylchloroform has been detected along with other halocarbons at various locations throughout the U.S. and around the world. The most extensive data are reproduced in Tables 2.1 and 2.2. These data are taken from a study done at Cook College, Rutgers University (Lillian et al., 1975). Other data are summarized in Table 2.3. In addition, various industrial sites were monitored for methylchloroform in a study carried out by the Battelle Columbus Laboratories in late 1976 and early 1977 (see the last section of this chapter, "Methylchloroform Near Industrial Sites--Multimedia Levels").

The concentration of methylchloroform in the atmosphere ranges from about 0.1 $\mu g/m^3$ (20 ppt) in remote areas to over 500 $\mu g/m^3$ (100 ppb) in areas where the substance is manufactured or used. Pearson and McConnell (1975) point out that as one moves away from a manufacturing facility, the concentration of methylchloroform in air drops off rapidly (Table 2.3). Battelle data support this conclusion and Ohta et al. (1976) make a similar observation. They state that the distribution peak for methylchloroform coincides with locations of machine or metal products plants which use the solvent.

It was found in the Battelle study that the highest concentrations of methylchloroform are generally observed downwind from a producer or user site and the concentration seems to be dependent on the distance from the discharge point. Most of the higher concentrations are observed at distances of less than 1 km. Considerable variation, however, was observed in the maximum downwind levels of methylchloroform at various production sites. The variations in the observed maximum concentration among plants may be due to differences in (1) production processes, (2) emission control equipment, (3) meteorological conditions, and (4) distance from the plant. Higher production capacity apparently does not necessarily imply higher emissions since the maximum concentrations observed at the larger plants were no higher than those observed at the smaller operations, and were sometimes lower. Large temporal variations are observed when measuring these chlorinated hydrocarbons downwind from a production facility.

TABLE 2.1. MAXIMUM AND MINIMUM LEVELS OF METHYLCHLOROFORM IN THE ATMOSPHERE AT VARIOUS LOCATIONS IN THE UNITED STATES

Monitoring Period and Location	Levels	Concentration, ppb
June 18-19, 1974	Maximum	0.20
Seagrit, New Jersey	Minimum	0.044
(National Guard Base)	Mean	0.10
June 27-28, 1974	Maximum	1.6
New York, New York	Minimum	0.10
(45th and Lexington)	Mean	0.61
July 2-5, 1974	Maximum	0.33
Sandy Hook, New Jersey	Minimum	0.030
(Fort Hancock)	Mean	0.15
July 8-10, 1974	Maximum	0.30
Delaware City, Delaware	Minimum	0.03
(Road 448 and Route 72 intersection)	Mean	0.10
July 11-12, 1974	Maximum	0.21
Baltimore, Maryland	Minimum	0.044
(1701 Poncabird Pass, Ford Holabird area)	Mean	0.12
July 16-26, 1974	Maximum	0.35
Wilmington, Ohio	Minimum	0.030
(Clinton County Air Force Base)	Mean	0.097
September 16-19, 1974	Maximum	0.13
White Face Mountains	Minimum	0.032
(New York State)	Mean	0.067
March-December, 1973	Maximum	14.4
Bayonne, New Jersey	Minimum	0.075
	Mean	1.59

Source: Lillian et al., 1975.

TABLE 2.2. TYPICAL LEVELS OF METHYLCHLOROFORM IN THE ATMOSPHERE

Date and Time	Location	Methylchloroform Concentration, ppb
June 27, 1974 2300	New York, New York Urban	0.28
September 17, 1974 1200	White Face Mountains New York State (nonurban)	0.083
July 2, 1974 1400	Over Ocean Sandy Hook, New Jersey 4.8 km (3 mi) offshore)	0.18
July 19, 1974 1300	Seagirt, New Jersey (National Guard Base)	0.072
July 17, 1974	Above the Inversion elevated 1500 m (5000 ft) Wilmington, Ohio	0.025
July 17, 1974 1203	Inversion Layer elevation 450 m (1500 ft) Wilmington, Ohio	0.065

Source: Lillian et al., 1975.

TABLE 2.3. MISCELLANEOUS MONITORING DATA FOR METHYLCHLOROFORM IN THE ATMOSPHERE

Location	Date of Data Collection	Concentration	Method[a]	Reference
Los Angeles Basin	Fall 1972	0.37 ppb (avg.)	GC/EC	Simmonds et al., 1974
San Bernadino Mts.	Fall 1972	0.05 ppb (avg.)	"	" "
New Brunswick NJ	1973	0.27 ppb	Coulometric GC	Lillian and Singh, 1974
"	Unreported	0.83 ppb	"	" "
Kansas City-NASN Station	1974	Detected	GC/MS	Bunn et al., 1975
Worldwide	1974	5×10^{-9} ml/ml of air	Estimate	Goldberg, 1975
Houston TX and vicinity	Nov. 1974	Detected	GC/MS computer	Pellizzari et al., 1976
Los Angeles Basin	April 1975	"	"	" "
Pullman WA	Dec. 1974 to Feb. 1975	100±15 ppt	GC/MS	Grimsrud and Rasmussen, 1975
Western Ireland	June/July 1974	64.8 ppt	Coulometric GC	Lovelock, 1974
North Atlantic	Oct. 1973	75.1 ppt	"	"
Britain, perimeter of a manufacturing plant	1972-74	16 ppb (mass)	GC/EC	Pearson and McConnell, 1975
Heath, near the above plant	1972-74	6.2 to 11 ppb (mass)	"	" "
Suburban area, removed from plant	1972-74	<0.1 to 6 ppb (mass)	"	" "
Tokyo	May 1974-April 1975	0.8 ppb (annual avg.)	"	Ohta et al., 1976
Southern Hemisphere	1974	24.4 ppt	"	Cox et al., 1976
Northern Hemisphere	1974	64.8 ppt	"	" "
Stanford Hills CA	Nov. 1975	77.6 ppt (avg. 75 measurements)	"	Singh et al., 1977
Point Reyes CA	Dec. 1975	90.3 ppt (avg. 300 measurements)		" "

[a] GC/EC = Gas chromatography with electron capture detector; GC/MS = Gas chromatography, mass spectroscopy

Changes in meteorological conditions, particularly wind speed and direction, and/or variations in the emissions may account for this phenomenon.

There is some question about the source of methylchloroform in the atmosphere. Lovelock (1974) suggested that some methylchloroform may occur naturally while discussing evidence for the natural origin of carbon tetrachloride. He suggests that carbon tetrachloride may result from a biological source such as algae or more probably from an atmospheric process. He cites the following reasons for postulating a natural origin of carbon tetrachloride: (1) the abundance of carbon tetrachloride does not differ appreciably between the northern and southern hemispheres, which is not consistent with a northern industrial source, (2) air arriving at Western Ireland shows a strong correlation between the concentration of fluorocarbons and continental European origin. No such correlation is observed for carbon tetrachloride, (3) reaction in air between methane and chlorine at a concentration of 10^{-4} results in the production of small but significant amounts of carbon tetrachloride in the laboratory. After this discussion, Lovelock concludes that "there is some indication from their spatial distribution that chloroform and methylchloroform may also in part at least have a similar natural origin to carbon tetrachloride".

More recently, Cox et al. (1976), in considering oxidation of methylchloroform and other halocarbons by OH radicals, came to the conclusion that the industrial output of methylene chloride, chloroform, and methylchloroform is probably insufficient to balance the sink due to OH attack, pointing to a natural source. These workers point out, however, that the observed hemispheric concentration differences are consistent with an anthropogenic source (see Table 2.3).

The natural source theory for carbon tetrachloride has recently come under attack. Galbalby (1976) concludes that a large fraction, perhaps all of the carbon tetrachloride observed in the atmosphere, could be man-made, and carbon tetrachloride is a global atmospheric pollutant. He attributes the ubiquitous nature of carbon tetrachloride and the similar concentrations found in background air in both hemispheres to the fact that carbon tetrachloride has a long lifetime in the atmosphere, and that a near equilibrium state of man-made emissions and destruction of the compound in the atmosphere exists. If this is true, it seems unlikely that a more complicated C_2 molecule such as methylchloroform could be generated in the atmosphere in any quantity.

From chemical kinetic computations, Graedel and Allara (1976) conclude that the possibility is remote that any of the observed atmospheric halocarbons including methylchloroform and carbon tetrachloride are produced from natural or anthropogenic precursors by atmospheric chemical processes.

Based on their measured background concentration for methylchloroform of 84 ppt in the northern hemisphere (Table 2.3) and on a calculated cumulative worldwide emissions output of 3.3 million tons of methylchloroform up to December, 1975, which they calculate would correspond to a background atmospheric concentration of 146 ppt, Singh et al. (1977)

conclude that no natural sources for methylchloroform can be inferred. However, they suggest that methylchloroform residence time in the troposphere must be much longer than the 1.1 years suggested by others (Table 3.1) or else secondary anthropogenic or natural sources must exist.

The resolution of this question depends on the gathering of more data and a better understanding of the atmospheric chemistry of halocarbons.

METHYLCHLOROFORM IN SOIL AND SEDIMENT

The only information available on methylchloroform levels in soil and sediment was obtained from the Battelle study (Battelle's Columbus Laboratories, 1977). This information is presented in the section on Methylchloroform Near Industrial Sites.

In general, the concentrations in soil range from less than 0.1 ppb to about 1 ppb. There does not seem to be any correlation with the distance from production or user sources, and a concentration of 0.42 ppb was found in a background sample taken many miles from any known source of methylchloroform.

Methylchloroform levels in sediment samples were somewhat higher on the average than the levels in soil. Levels in sediment ranged from less than 0.1 ppb to about 6 ppb. The background level was 0.45 ppb at a site far removed from known sources of methylchloroform.

METHYLCHLOROFORM IN SURFACE WATERS

Approximately 200 water samples have been collected and analyzed for various organic substances (Chian and Ewing, 1976). These samples were collected from 14 heavily industrialized river basins. These areas and the approximate number of samples taken at each location are indicated in Figure 2.1 (Chian and Ewing, 1976, Progress Report No. 4). The results are summarized in Table 2.4. Methylchloroform was detected in 63 of the approximately 200 samples analysed and the concentrations ranged from less than 1 ppb to 8 ppb in these surface waters.

In the vicinity of production plants, the concentration of methylchloroform in surface waters is much higher. Levels up to 200 ppb are common and at one site a level of 3.3 ppm was found. The data appear in the last section of this chapter.

Pearson and McConnell (1975) report methylchloroform concentrations of 0.09 ppb in rainwater collected in Runcorn, England. The highest concentrations that these researchers measured in upland river waters was 0.3 ppb. These same authors also reported that they have never detected organochlorines in well waters. With a normal detection limit of 0.2 ppb, Pearson and McConnell (1975), between April and August, 1973, determined that the maximum concentration of methylchloroform in Liverpool Bay

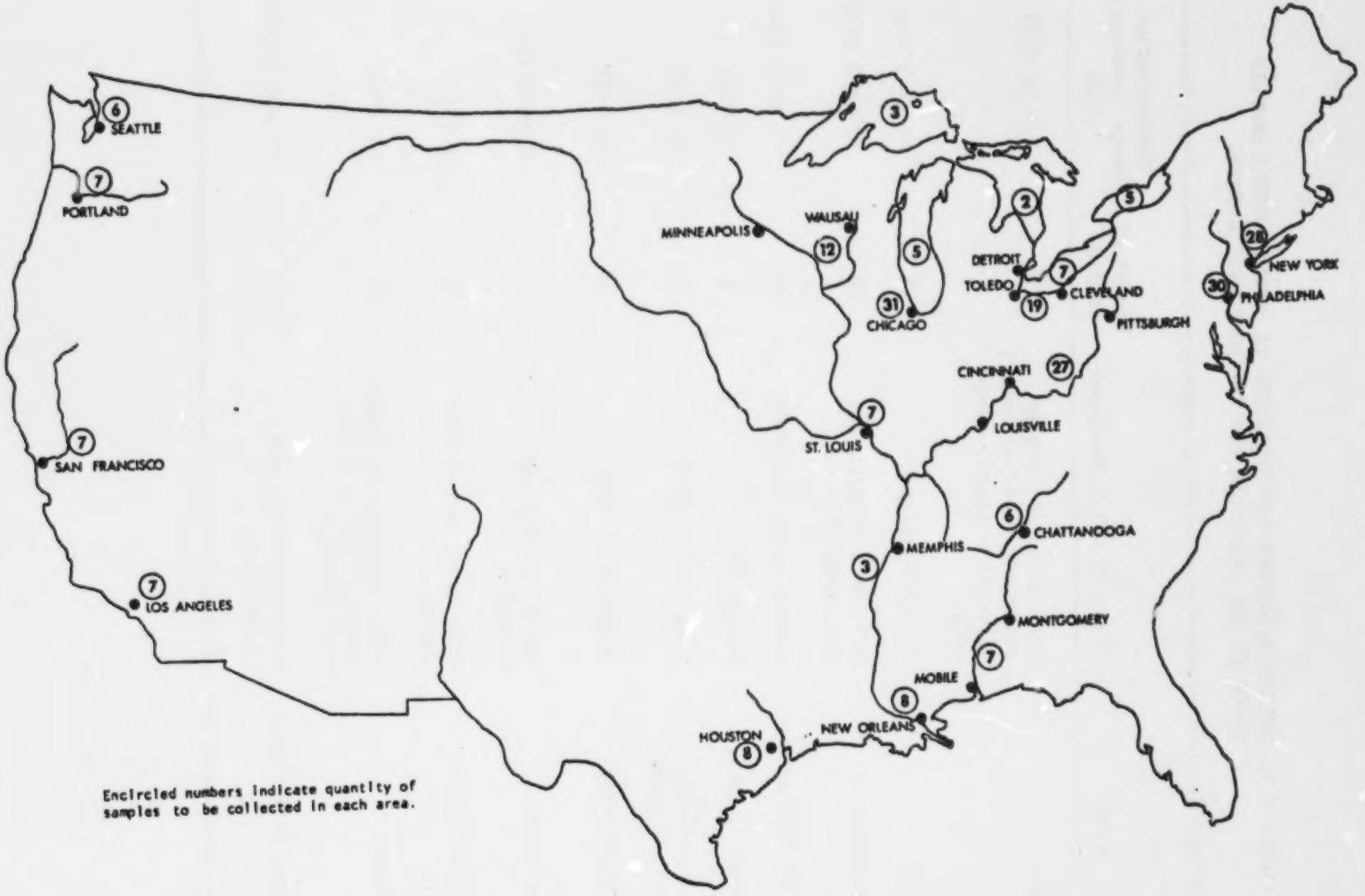


Figure 2.1. Industrialized areas where surface water was sampled (Source: Chian and Ewing, 1976).

TABLE 2.4. METHYLCHLOROFORM CONCENTRATION IN SURFACE WATER SAMPLES TAKEN BY THE INSTITUTE FOR ENVIRONMENTAL STUDIES

Area	Type of Water Analyzed	Number of Samples	Concentration Range (Average), ppb
Chicago	Lake Michigan, sewage treatment plant effluent, filtration plant, channels	7	0.5 to 8(3)
Illinois	Illinois River	11	<1 to 3 (<1)
Pennsylvania	Delaware, Schuylkill, and Lehigh Rivers	12	<1 to 3 (<1)
New York City area	Hudson River and bays	14	<1 to 2 (<1)
Hudson River area	Hudson River	1	<1 (<1)
Upper and Middle Mississippi River	Mississippi River	3	<1 (<1)
Lower Mississippi River	Mississippi River	1	<1 (<1)
Houston area	Galveston Bay and channels	3	1 to 2 (1)
Ohio River Basin	Ohio River and tributaries	3	<1 (<1)
Great Lakes	Lake Superior, Michigan, Huron, Ontario, Erie, and vicinity	6	<1 (<1)
Tennessee River Basin	Tennessee River and tributaries	2	<1 and 4 (<2)

seawater was 3.3 ppb. In Liverpool Bay sediments, a maximum combined concentration of methylchloroform and carbon tetrachloride was 9.9 ppb.

METHYLCHLOROFORM IN DRINKING WATER

The National Organics Reconnaissance Survey (NORS) was initiated by the U.S. Environmental Protection Agency in November, 1974. NORS had three major objectives: (1) Determine the extent of the presence of four trihalomethanes in finished water; (2) Determine what effects the source and treatment of water had on the formation of these compounds; (3) Characterize as completely as possible the organic content of 10 drinking waters from sites representing five major categories of raw water souces.

During the survey of 10 cities, at least 129 compounds were identified in drinking water. Some were quantified and others were detected without quantification. Methylchloroform was one of the latter. Its presence was detected in the drinking water in Ottumwa, Iowa; Philadelphia, Pennsylvania; and Cincinnati, Ohio (U.S. Environmental Protection Agency, 1975b). Methylchloroform was also detected in the drinking water from the Belmont water treatment plant in Philadelphia, Pennsylvania, on August 8, 1975, using continuous liquid-liquid extraction of the water followed by identification by gas chromatography/mas spectrometry (Keith, 1976). Methylchloroform was also detected in tap water at the National Institute of Environmental Health Sciences, Research Triangle Park, North Carolina, on May 7, 1975, using gas chromatography/mass spectrometry (Keith, 1976).

Methylchloroform was identified as a component of New Orleans drinking water but it was not quantified (Dowty et al., 1975a). The only quantitative information available was that reported by Bellar et al. (1974). In an investigation of the chlorination of water for purification and the potential for the formation of potentially harmful chlorinated compounds by the process, scientists at the National Environmental Research Center at the Environmental Protection Agency in Cincinnati, Ohio, reported the following concentrations of methylchloroform in water from a sewage-treatment plant: influent before treatment, 16.5 µg/ℓ; effluent before chlorination, 9.0 µg/ℓ; and effluent after chlorination, 8.5 µg/ℓ.

METHYLCHLOROFORM NEAR INDUSTRIAL SITES--MULTIMEDIA LEVELS

A program to determine environmental levels of methylchloroform was initiated in 1976 at Battelle's Columbus Laboratories. During late 1976 and early 1977, samples were collected from various production sites, a user site, and a background site. The samples were analyzed and the results are summarized in Tables 2.5 through 2.10 and in the corresponding maps of the plant locations on which the sampling sites are indicated Figures 2.2 through 2.7). Details of the results and methodology are given in a companion report, EPA-560/6-77-025 (Battelle's Columbus Laboratories, 1977).

TABLE 2.5. CONCENTRATION OF METHYLCHLOROFORM IN AIR, WATER, SOIL, AND SEDIMENT AT DOW CHEMICAL PLANT A (METHYLCHLOROFORM PRODUCER)

Air

Site	Concentration, ppbv[a]	Distance From Plant, km	Upwind (U), Downwind (D), or Variable (V)
1A	<0.3 to 2.3	2.6	D
2A	<0.3 to 1.1	1.9	V
3A	<0.3 to 1.2	3.2	D
4A	<0.3	2.1	D
5A	<0.3 to 0.7	0.8	U
6A	<0.3	1.9	U
7A	<0.3 to 1.1	2.6	V
8A	<0.3 to 2.2	2.6	D
10A	<0.3	3.2	D
12A	<0.3 to 11.5	2.6	D
13A	<0.3 to 0.5	5.1	D
14A	<0.3	7.8	D

Water, Soil, and Sediment

Site	Description of Media	Concentration, ppb
A1	Surface water, 10 m upstream of effluent canal	117
A2	Water, as above, except 2.5 m deep	119
A5	Surface water, 400 m downstream of plant outfall	0.8
A6	Water, as above except 4 m deep	1.0
A7	Surface water, 800 m upstream of plant outfall	0.1
A8	Surface water, bayou 2.6 km from plant	12
A2,A4, A7,A12	Soil, quadrants around plant at 2 km	0.06 to 0.68
A1S	Sediment, from effluent canal	6.1
A5S	Sediment, 400 m downstream of plant outfall	0.34
A7S	Sediment, 800 m upstream of plant outfall	0.31

[a]Limits of detection: 0.3 ppbv. To convert to $\mu g/m^3$ at 25 C, multiply ppbv by 5.46.

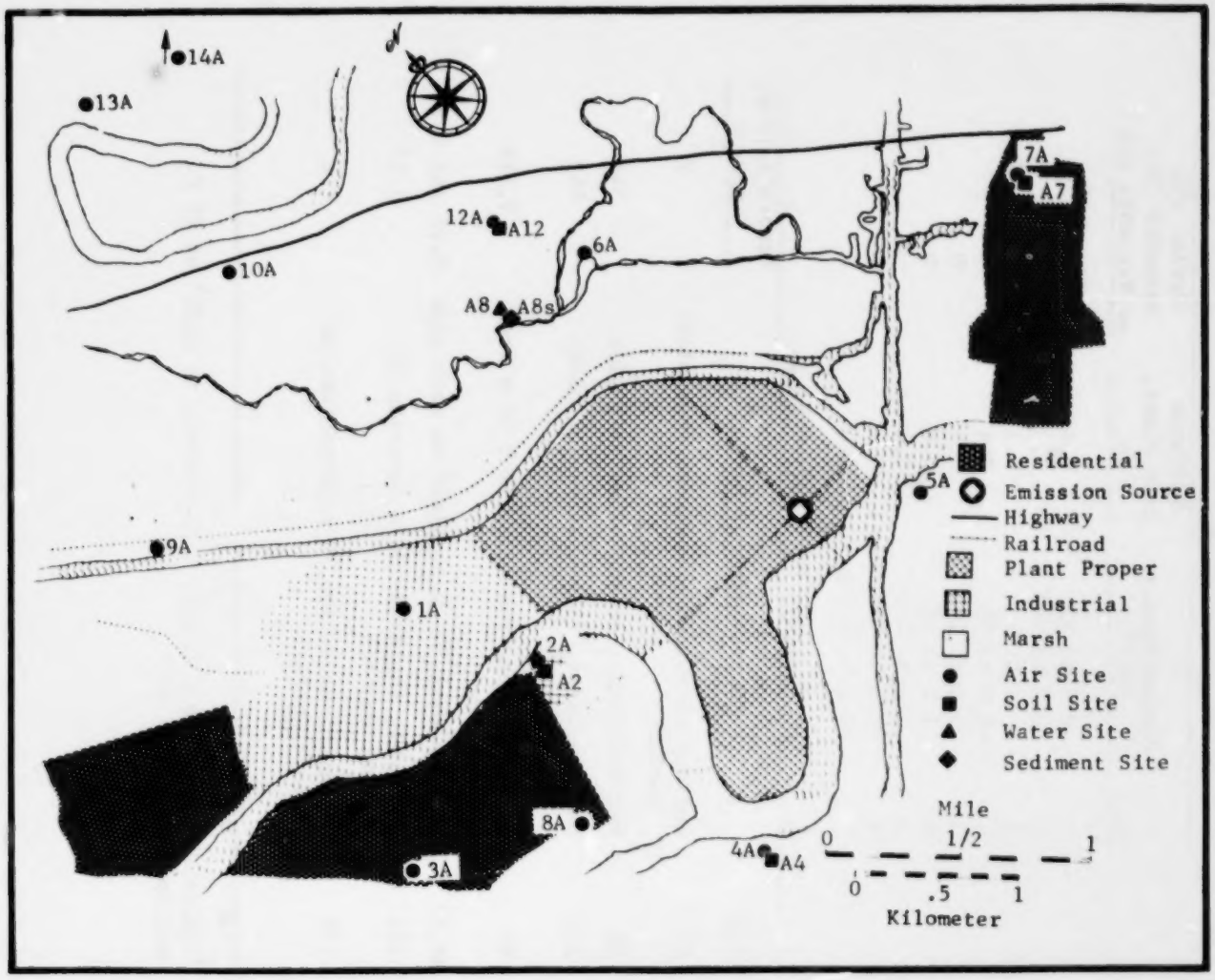


Figure 2.2. Sampling locations at Dow Chemical Plant A, Freeport, texas--methylchloroform production site.

TABLE 2.6. CONCENTRATION OF METHYLCHLOROFORM IN AIR, WATER, SOIL, AND SEDIMENT AT VULCAN MATERIALS COMPANY (METHYLCHLOROFORM PRODUCER)

Air

Site	Concentration, ppbv[a]	Distance From Plant, km	Upwind (U), Downwind (D), or Variable (V)
1	<0.3 to 18	0.4	V
2	<0.3 to 140	0.3	V
3	<0.3	0.3	V
4	<0.3 to 75	0.4	D
5	5.5 to 155	0.6	D
6	1.2 to 14	0.6	D
7	1.4 to 4.0	0.4	-
8	<0.3 to 0.8	0.3	V
9	<0.3 to 0.5	3.0	D

Water, Soil, and Sediment

Site	Description of Media	Concentration, ppb
E1	Surface water, 30 m upstream from plant outfall	2
E2	Surface water, at end of outfall pipe	344
E3	Surface water, 75 m upstream of plant outfall	169
E4	Surface water, roadside ditch 60 m from plant	3,314
E6,E7	Soil, within 200 m of plant on each side	0.45 and 0.94
E1S	Sediment, shoreline 30 m upstream of plant outfall	0.13
E3S	Sediment, shoreline 75 m downstream of plant outfall	2.6

[a]Limits of detection 0.3 ppbv. To convert to $\mu g/m^3$ at 25 C, multiply ppbv by 5.46.

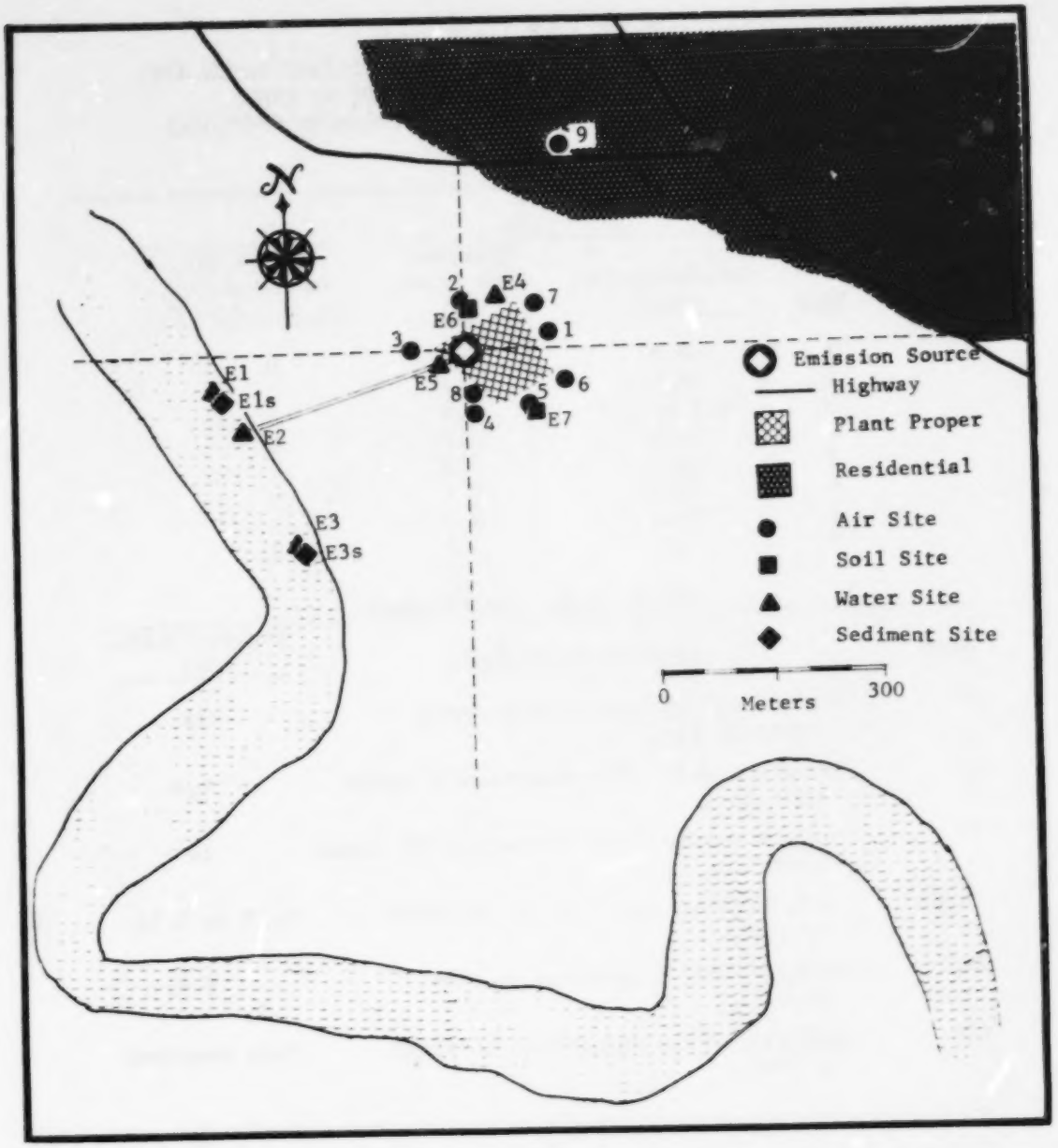


Figure 2.3. Sampling locations at Vulcan Materials Company, Geismar, Louisiana--methylchloroform production site.

TABLE 2.7. CONCENTRATION OF METHYLCHLOROFORM IN AIR, WATER, SOIL, AND SEDIMENT AT ETHYL CORPORATION (METHYLCHLOROFORM PRODUCER)

Air			
Site	Concentration, ppbv[a]	Distance From Plant, km	Upwind (U), Downwind (D), or Variable (V)
1	<0.3	0.4	D
2	<0.3	0.2	D
3	<0.3 to 3.9	2.4	D
4	<0.3	2.6	D
5	<0.3	2.2	-
6	<0.3	0.7	U
7	<0.3	2.2	-
8	<0.3	3.2	D

Water, Soil, and Sediment		
Site	Description of Media	Concentration, ppb
C1	Surface water, immediately above settling pond	74
C2	Surface water, 200 m upstream of plant outfall	9,4
C3	Surface water, 300 m downstream of plant outfall	20
C4,C7	Soil, various locations in vicinity of plant	0.13 to 0.28
C2S	Sediment, 200 m upstream of plant outfall	0.81
C3S	Sediment, 300 m downstream of plant outfall	None detected

[a]Limits of detection: 0.3 ppbv. To convert to $\mu g/m^3$ at 25 C, multiply ppbv by 5.46.

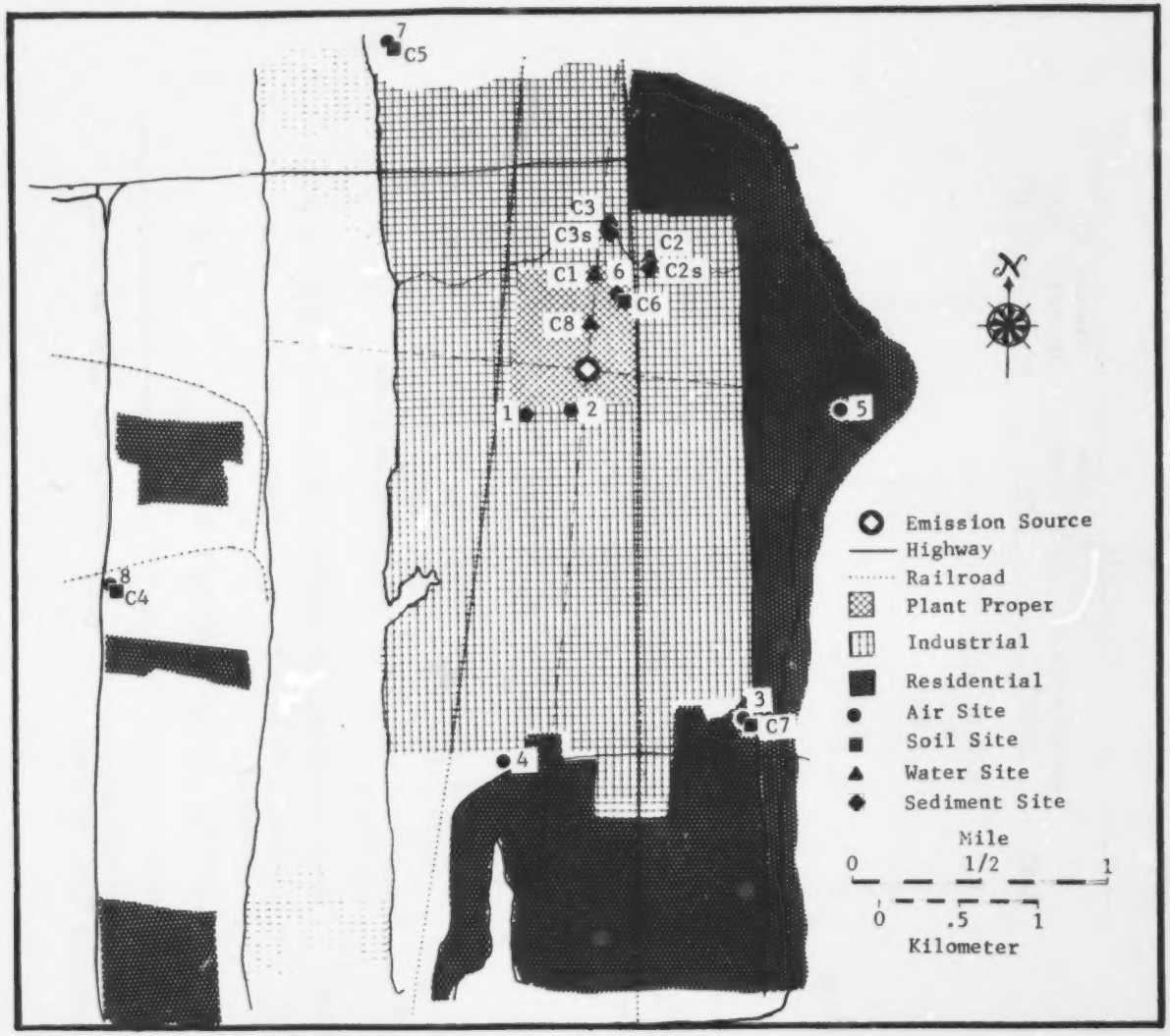


Figure 2.4. Sampling locations at Ethyl Corporation, Baton Rouge, Louisiana--methylchloroform production site.

TABLE 2.8. CONCENTRATION OF METHYLCHLOROFORM IN AIR, WATER, SOIL, AND SEDIMENT AT PPG INDUSTRIES (METHYLCHLOROFORM PRODUCER)

Air

Site	Concentration, ppbv[a]	Distance From Plant, km	Upwind (U) Downwind (D), or Variable (V)
1	0.8 to 1.3	1.3	U
2	<0.3	4.2	U
3	<0.3 to 0.7	3.5	V
4	<0.3	2.7	U
5	<0.3	1.4	D
6	<0.3 to 8.5	4.0	D
7	<0.3	0.6	U
8	0.4	1.3	U

Water, Soil, and Sediment

Site	Description of Media	Concentration, ppb
F1	Surface water, 50 m upstream of plant outfall	132
F2	Surface water, at plant outfall No. 1	181
F3	Surface water, at plant outfall No. 2	58
F4	Surface water, 50 m downstream of outfall No. 2	161
F5	Surface water, lake downstream of plant outfall	5
F6-F9	Soil, quadrants surrounding plant	0.14 to 1.0
F1S	Sediment, 50 m upstream of plant outfall	2.2
F3S	Sediment, at plant outfall No. 2	1.1

[a]Limits of detection: 0.3 ppbv. To convert to $\mu g/m^3$ at 25 C, multiply ppbv by 5.46.

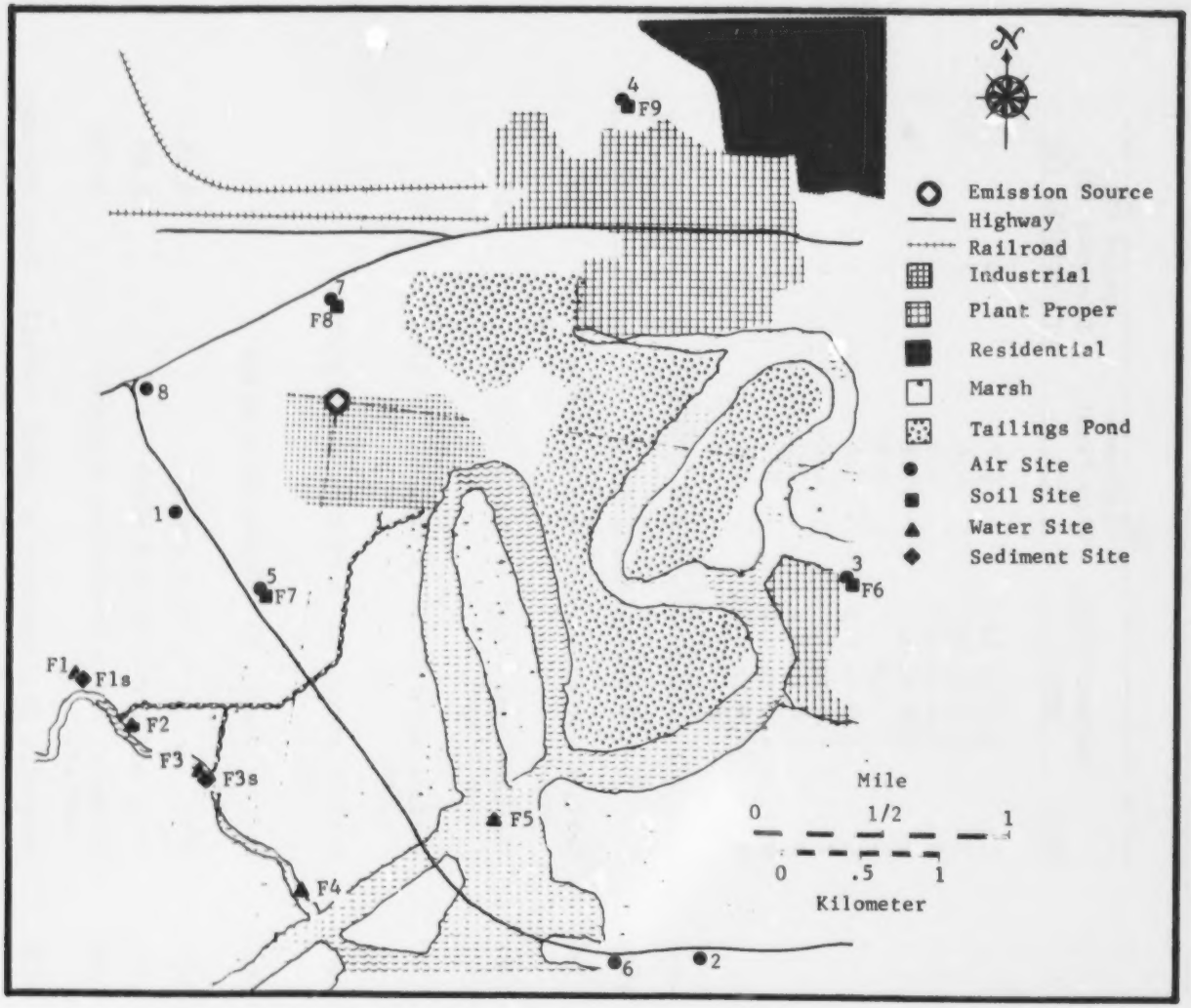


Figure 2.5. Sampling locations at PPG Industries, Lake Charles, Louisiana--methylchloroform production site.

TABLE 2.9. CONCENTRATION OF METHYLCHLOROFORM IN AIR, WATER, SOIL, AND SEDIMENT AT BOEING COMPANY (METHYLCHLOROFORM USER)

Air			
Site	Concentration, ppbv[a]	Distance From Plant, km	Upwind (U) Downwind (D), or Variable (V)
1	0.8 to 10.0	0.7	D
2	0.4 to 0.5	0.6	U
3	0.4 to 5.0	0.9	U
4	0.6 to 6.2	1.1	-
5	0.9	0.9	-
6	1.6 to 2.3	0.4	-
7	4.8 to 7.4	1.2	D
8	7.8 to 8.4	2.0	-
9	4.0 to 4.4	2.9	U
10	7.0 to 8.1	1.1	-

Water, Soil, and Sediment		
Site	Description of Media	Concentration, ppb
J2	Surface water, outfall from settling pond	18
J6	Surface water, outfall canal, 1.5 km from plant	18
J3	Surface water, outfall canal, 3 km from plant	12
J4	Surface water, 100 m downstream of plant outfall	6
J5	Surface water, 30 m upstream of plant outfall	6
J1	Soil, about 1 km from plant	0.40
J7	Soil, about 0.5 km from plant	0.65
J4S	Sediment, 100 m downstream of plant outfall	0.039
J5S	Sediment, 30 m upstream of plant outfall	None detected

[a]Limits of detection: 0.3 ppbv. To convert to $\mu g/m^3$ at 25 C, multiply ppbv by 5.46.

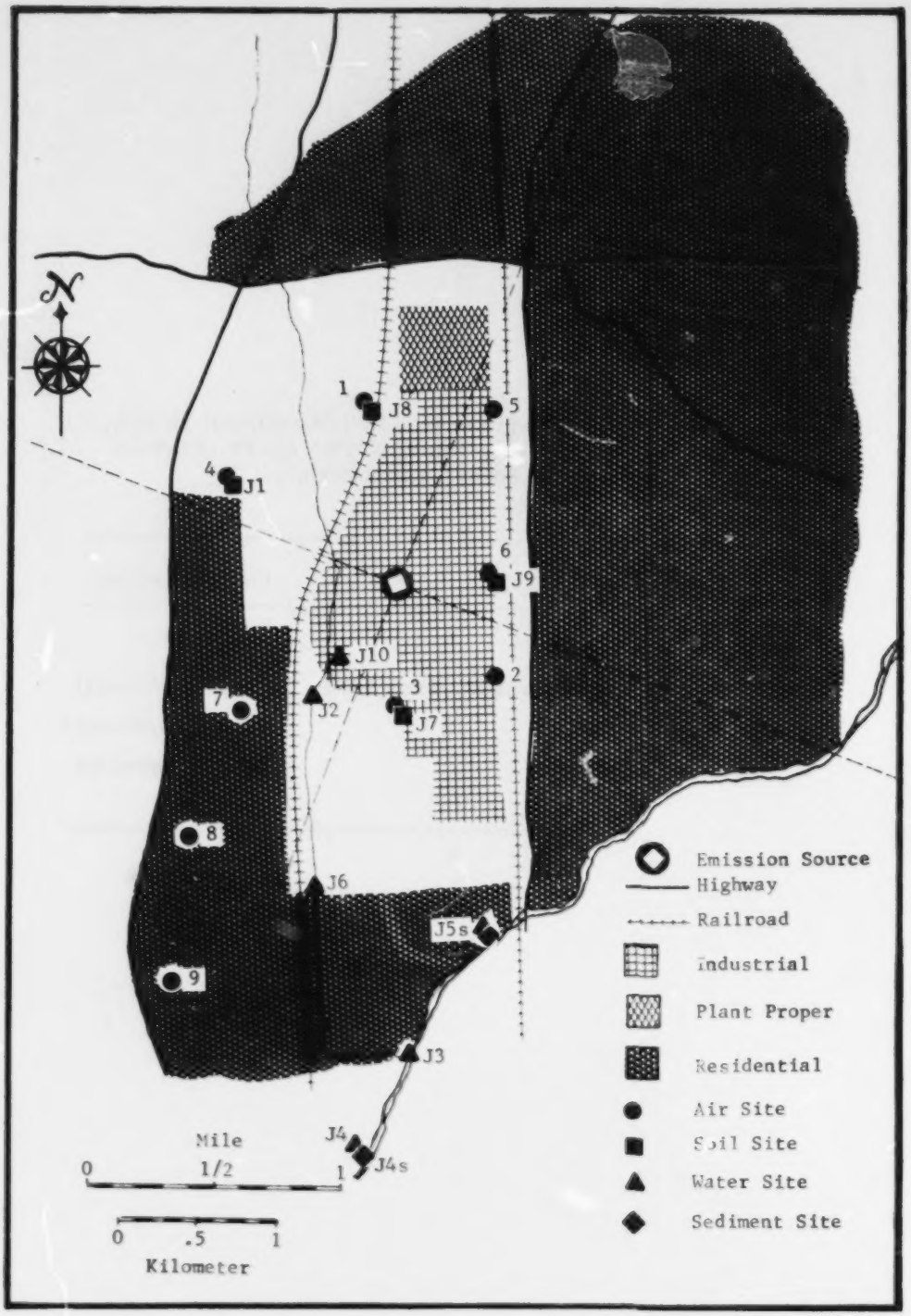


Figure 2.6. Sampling locations at Boeing Company, Auburn, Washington--methylchloroform user site.

TABLE 2.10. CONCENTRATION OF METHYLCHLOROFORM IN AIR, WATER, SOIL, AND SEDIMENT AT ST. FRANCIS NATIONAL FOREST (BACKGROUND)

Media	Concentration
Air	<0.3 ppbv
Surface water, from lake	0.4 ppb (average)
Soil	0.42 ppb (average)
Sediment	0.45 ppb (average

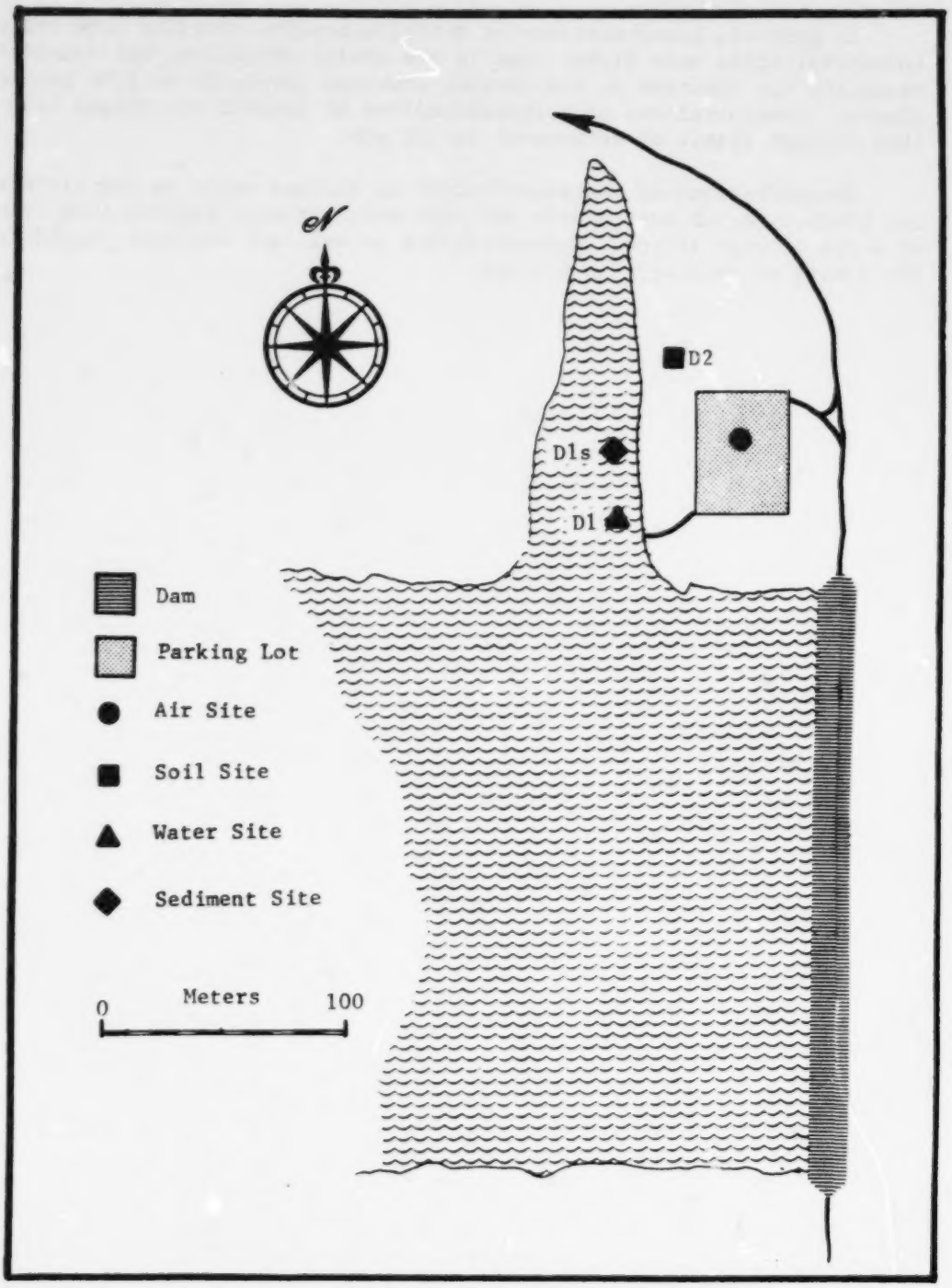


Figure 2.7. Sampling locations at St. Francis National Forest, Helena, Arkansas--background site.

In general, concentrations of methylchloroform downwind from the various industrial sites were higher than in the upwind direction, but considerable variation was observed in the maximum downwind levels at various production plants. Concentrations of methylchloroform in ambient air ranged from less than 0.3 ppb (limit of detection) to 155 ppb.

Concentrations of methylchloroform in surface water in the vicinity of the production and user plants was even more variable ranging from fractions of a ppb to over 16 ppm. Concentrations in soil and sediment ranged from the limits of detection to 6.1 ppb.

3. TRANSFORMATIONS OF METHYLCHLOROFORM IN THE ENVIRONMENT

This section indicates the changes that methylchloroform can undergo in various real and simulated environmental media. The information on the subject is summarized in Table 3.1 and represented schematically in Figure 3.1.

Methylchloroform is oxidized photochemically in the troposphere. The photochemical removal rate is estimated to be 0.9 million tons per year (Cox et al., 1976), which is in excess of world production resulting in release estimated by the same authors to be 0.28 millions tons per year. The implications of these results are discussed in Chapter 2. The residence of methylchloroform in the troposphere is longer than that for a compound containing unsaturation such as trichloroethylene, but much shorter than the residence time for completely halogenated compounds such as carbon tetrachloride or the freons with lifetimes of over 300 years.

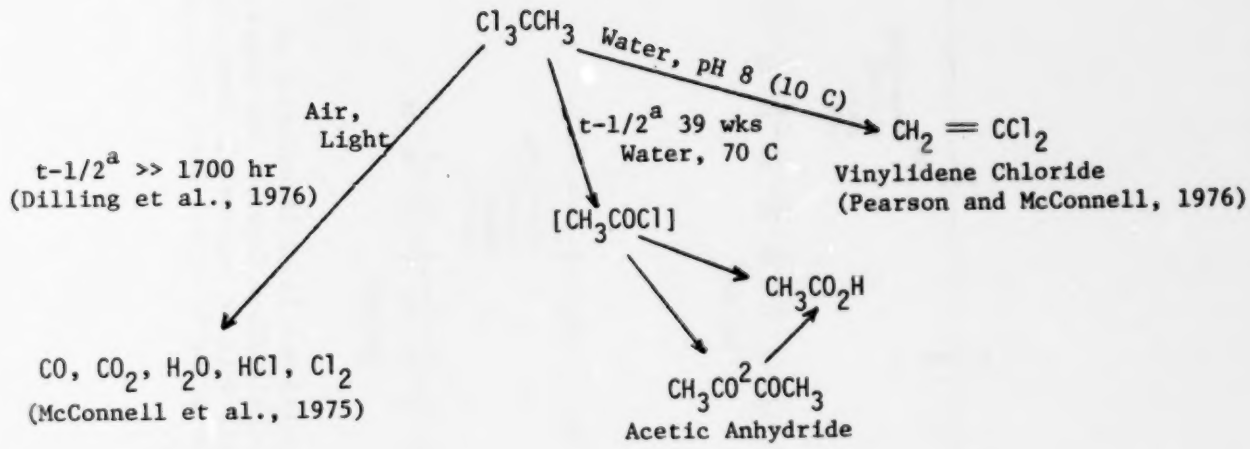
There is evidence that methylchloroform is relatively stable under simulated sea-level conditions as shown in Figure 3.2 Sunlight in these experiments was initiated using sunlamps with an intensity of one to two times that of noontime sea-level sunlight at wavelengths greater than 295 nm. Figure 3.3 shows the rapid oxidation of methylchloroform at simulated high-altitude conditions. These conditions, however, are some 10 to 100 times as intense as would be observed from 40 km upward, and so the rate is probably exaggerated. These results indicate that methylchloroform is fairly nonreactive in sea-level atmospheres but as it mixes upward its photochemical decomposition increases.

Methylchloroform has a half-life of 30 weeks in sea water of pH 8 at 10 C. The primary degradation product is vinylidene chloride (Pearson and McConnell, 1975). However, at higher temperatures (approximately 70 C), methylchloroform hydrolyses to acetic acid. Evaporation from water into the atmosphere is probably the principal mode of removal. Dilling et al. (1975) estimate that a half-life of 17 minutes for evaporation. In good agreement is the data from Dow Chemical, Table 3.2. Here evaporation from a variety of substances is reported.

Pearson and McConnell (1975) were unable to identify any biological oxygen demand (BOD) for the chlorinated hydrocarbons.

TABLE 3.1. TRANSFORMATIONS OF METHYLCHLOROFORM IN THE ENVIRONMENT

Media	Change or Products Observed	Reference
Simulated atmospheric conditions	<5% decomposed in 23.5 hr with (NO)	Dilling, et al., 1976
Simulated atmospheric conditions	<5% decomposed in 8 hr (50 ppm methylchloroform 10 ppm NO_2)	Dilling, et al., 1976
Simulated atmospheric conditions	Est. half-life >>1700 hr	Dilling, et al., 1976
One ppm in water containing natural and added contaminants	17 min half-life for evaporation	Dilling, et al., 1975
Water, 25C	6.9 mo half-life for hydrolysis	Dilling, et al., 1975
Sea Water (pH 8, 10 C)	9 mo half-life, vinylidene chloride and acetic acid	McConnell, et al., 1975
Atmosphere	10-33 weeks half life	McConnell, et al., 1975
Troposphere	1.1 years (lifetime)	Cox, et al., 1976
Troposphere, 4.4 parts/thousand	26 weeks half life	Pearson and McConnell, 1975
Smog chamber	Ozone	Farber, 1973
Atmosphere near welding	HCl, Cl_2 (in low concentrations)	Rinzema and Silverstein, 1972



[a]t-1/2 = time required for one-half of the chlorinated hydrocarbon to disappear by the indicated process.

Figure 3.1. Transformations of methylchloroform.

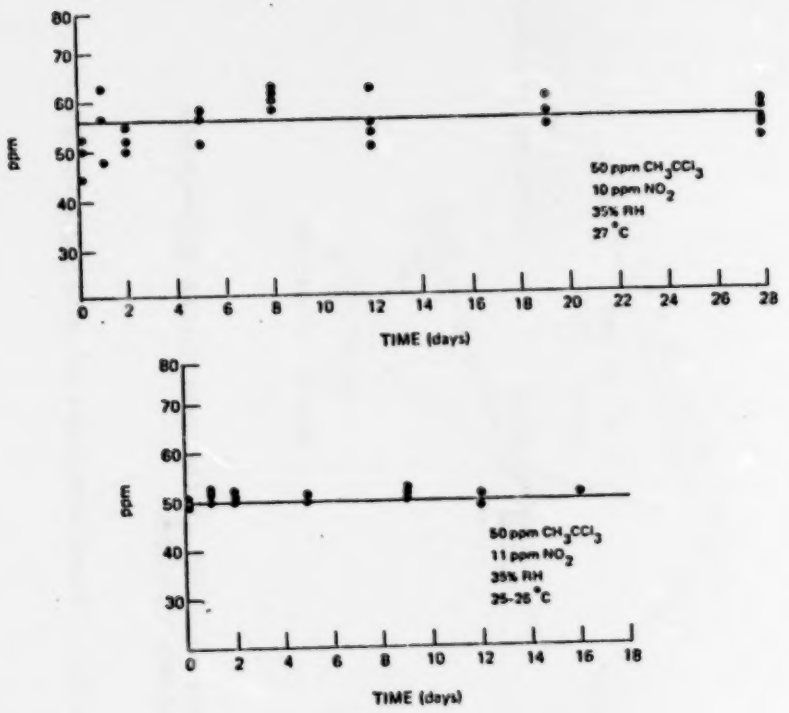


Figure 3.2. Simulated sea level irradiation of methylchloroform (Source: Dow Chemical Company data as reported by Study Panel on Assessing Potential Ocean Pollutants, 1975).

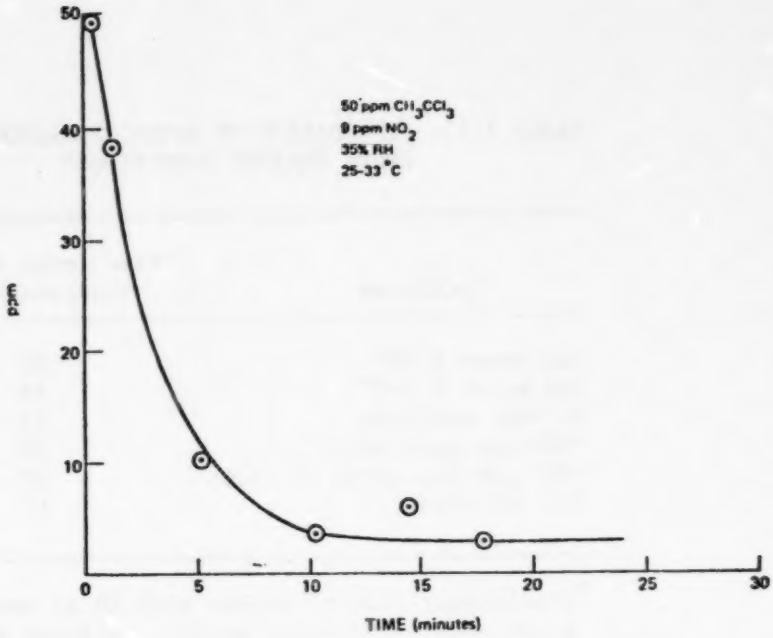


Figure 3.3. High altitude photoreaction of methylchloroform (Source: Dow Chemical Company data as reported by Study Panel on Assessing Potential Ocean Pollutants, 1975).

TABLE 3.2. EVAPORATION OF METHYLCHLOROFORM UNDER VARIOUS CONDITIONS[a]

Condition	Time (min) for 50% Disappearance
Tap water @ 25°	22
Tap water @ 1-2°	33
3% Salt solution	25
∿500 ppm peat moss	20
∿500 ppm wet bentonite clay	20
2.2 mph wind	17

[a]Conditions: 250 ml beaker with 20 ml water with 1 ppm solute initially at 25°. Stirred at 200 rpm.

Source: Dow Chemical Company data as reported by Study Panel on Assessing Potential Ocean Pollutants, 1975.

4. OCCURRENCE OF METHYLCHLOROFORM IN FOOD

There are very few data on the presence of methylchloroform in food raised and sold in the United States, but there is some information on the presence of methylchloroform in foodstuffs found in the United Kingdom. This information is summarized in Table 4.1. Methylchloroform concentrations on the order of parts per billion are found in several common foodstuffs.

TABLE 4.1. METHYLCHLOROFORM IN FOODSTUFFS

	Concentration, µg/kg
Meat	
English beef, steak	3
English beef, fat	6
Pig's liver	4
Oils and fats	
Olive oil (Spanish)	10
Cod liver oil	5
Castor oil	6
Fruits and vegetables	
Potatoes (S. Wales)	4
Potatoes (N.W. England)	1
Apples	3
Pears	2
Tea (packet)	7
Fresh bread	2

Source: McConnell et al., 1975.

5. EXPOSURE AND BIOLOGICAL ACCUMULATION OF METHYLCHLOROFORM IN MAN

EXPOSURE

NIOSH estimates that 100,000 workers are exposed to methylchloroform (U.S. National Institute for Occupational Safety and Health, 1976). Methylchloroform is the principal solvent in wet cleaning applications and its use as a vapor degreaser is expected to increase by 10 percent per year as trichloroethylene undergoes further restrictions (U.S. Environmental Protection Agency, 1975a).

A two-year series of studies involving cleaning operations throughout the United States was carried out by Dow Chemical (Skory et al., 1974). The purpose was to determine the extent of worker exposure during solvent vapor degreasing and to compare the three most commonly used chlorinated solvents: methylchloroform, trichloroethylene, and perchloroethylene. Dow estimates that there are over 25,000 chlorinated solvent vapor degreasers throughout the United States. The studies were conducted in the worker breathing zones which were adjacent to some 275 industrial vapor degreasing operations. The results of this study show that trichloroethylene and perchloroethylene vapor concentrations measured around vapor degreasers frequently exceeded the allowable standards for health and safety. Peak concentrations were high enough to present a definite health and safety hazard from anesthetic effects such as dizziness, lack of coordination, and impaired judgment. These authors further concluded that methylchloroform emissions during vapor degreasing can be controlled easily at levels below standards established by OSHA.

Although the national primary and secondary photochemical oxidant standards for chlorinated solvents are less than 3 lb/hour or 15 lb/day maximum for each equipment, it is not uncommon for an idling open-top vapor degreaser measuring 24 x 58 inches to lose 47 lb/day trichloroethylene or 33 lb/day methylchloroform (Archer, 1973). Judging from production figures, this material is being lost to the atmosphere and is then replaced.

It is estimated that 2×10^5 tons of chlorinated hydrocarbons are lost to the environment each year (Murray and Riley, 1973) and that 3×10^5 tons of methylchloroform are discharged annually (Shamel et al., 1975). In Los Angeles County alone, it is estimated that 500 tons/day of industrial effluents are released into the air, and of this amount, 25 tons are dry cleaning fluids and 95 tons are degreasing solvents, that is, chlorinated hydrocarbons (Simmonds et al., 1974).

BIOLOGICAL ACCUMULATION

Dowty et al. (1975b) in a paper on halogenated hydrocarbons in drinking water concludes that "in view of the lipophilic nature of halogenated hydrocarbons and their occurrence in drinking water, it is not surprising that they might be found accumulating in blood or other tissues". These authors, however, present no data or references to support their contentions.

A Study Panel on Assessing Potential Ocean Pollutants (1975) reports that the bioaccumulation of low-molecular weight chlorinated hydrocarbons is quite low compared to accumulation of chlorinated pesticides in vertebrates. This same group reports on another study in which it was determined that the bioaccumulation factor is determined by the partition of the compound between the water and the tissues of the organism, and further that the log of bioaccumulation is linearly related to the log of the partition coefficient between octanol and water for some compounds. This relationship offers a method of estimating bioaccumulation. Bioaccumulation for methylchloroform was estimated to be 13. This compound would be expected to act similarly to carbon tetrachloride in organisms, exhibiting rapid uptake to steady state concentration, and rapid clearance.

By far the most definitive study on bioaccumulation was carried out by Pearson and McConnell (1975. Some of the results that they report are complicated by the fact that methylchloroform and carbon tetrachloride were not distinguished in the early analytical work that was done. Nevertheless, these authors determined the amount of methylchloroform present in a large number of species and their results are tabulated in Table 5.1. They estimated that the maximum overall increase in concentration, between sea water and the tissues of animals at the top of the food chains such as fish liver, bird eggs, and seal blubber is less than 100-fold for solvents similar to methylchloroform; while a higher molecular weight chlorinated compound such as hexachlorobutadiene would have a maximum factor of 1000. They further concluded that the pattern of extensive bioaccumulation of marine food chains, which is postulated for chlorinated insecticides, does not appear. In laboratory tests where organisms are maintained for up to 3 months in apparatus similar to that used for toxicity determinations, Pearson and McConnell (1975) have shown that bioaccumulation can occur. Their results indicate the following: (1) the concentration of chlorinated hydrocarbons accumulated in a tissue tends to an asymptotic level, (2) concentrations in fatty tissues such as liver are higher than in muscle--concentration is proportional to fat content, and (3) when the test organism is returned to clean sea water, the concentration of the chlorinated hydrocarbon in the tissue falls. These researchers conclude that there is no evidence for the bioaccumulation of C_1/C_2 compounds in food chains and the maximum concentrations found in the higher trophic levels are still only parts per 10^8 by mass.

Despite this strong statement, it is based on a limited set of data, and caution should be exercised. More information is needed before final judgment is made about the accumulation of volatile chlorinated hydrocarbons in the tissues of animals and man.

TABLE 5.1. CHLORINATED HYDROCARBONS IN MARINE ORGANISMS
(concentrations expressed as parts per 10^9 by mass on wet tissue)

Species	Source	CCl_2CHCl	CCl_2CCl_2	$CH_3CCl_3+CCl_4$	
		Invertebrates			
Plankton	Liverpool Bay	0.05-0.4	0.05-0.5	0.03-10.7	0.04-0.9
Plankton	Torbay	0.0	2.3	2.2	
Nereis diversicolor (ragworm)	Mersey Estuary	ND	2.9	0.6	
Mytilus edulis (mussel)	Liverpool Bay	4-11.9	1.3-6.4	2.4-5.4	
	Firth of Forth	9	9	10	2
	Thames Estuary	8	1	5	0.7
Cerastoderma edule (cockle)	Liverpool Bay	6-11	2-3	0-2	0.4-1
Ostrea edulis (oyster)	Thames Estuary	2	0.5	0.9	0.1
Buccinum undatum (whelk)	Thames Estuary	ND	1	6	0.9
Crepidula fornicata (slipper limpot)	Thames Estuary	9	2	4	0.3
Cancer pagurus (crab)	Tees Bay	2.6	2.3	8.4	
	Liverpool Bay	10-12	8-9	5-34	3-5
	Firth of Forth	15	7	1	2
Carcinus maenas (shore crab)	Firth of Forth	12	6	14	3
Eupagurus bernhardus (hermit crab)	Firth of Forth	15	15	0.7	1
	Thames Estuary	5	2	2	0.2

TABLE 5.1. (Continued)

Species		Source	CCl_2CHCl	CCl_2CCl_2	CH_3CCl_3+CCl_4	
Crangon crangon (shrimp)		Firth of Forth	16	3	2	6
Asterias rubens (starfish)		Thames Estuary	5	1	5	0.8
Solaster sp. (sunstar)		Thames Estuary	2	2	3	0.2
Echinus esculentus (sea urchin)		Thames Estuary	1	1	3	0.1
			Marine algae			
Enteromorpha compressa		Mersey Estuary	19-20	14-14.5	24-27	
Ulva lactuca		Mersey Estuary	23	22	12	
Fucus vesiculosus		Mersey Estuary	17-18	13-20	9.4-10.5	
Fucus serratus		Mersey Estuary	22	15	35	
Fucus spiralis		Mersey Estuary	16	13	17	
			Fish			
Raja clavata (ray)	flesh	Liverpool Bay	0.8-5	0.3-8	2-13	
	liver	Liverpool Bay	5-56	14-41	1.5-18	
Pleuronectes platessa (plaice)	flesh	Liverpool Bay	0.8-8	4-8	0.7-7	
	liver	Liverpool Bay	16-20	11-28	2-47	

TABLE 5.1. (Continued)

Species		Source	CCL_2CHCl	CCl_2CCl_2	CH_3CCl_3+CCl_4	
Platycthys flesus	flesh	Liverpool Bay	3	2	4	2
(flounder)	liver	Liverpool Bay	2	1	3	0.3
Limanda limanda	flesh	Liverpool Bay	3-5	1.5-11	1.3-8	
(dab)	liver	Liverpool Bay	12-21	15-30	2-14	
Scomber scombrus	flesh	Liverpool Bay	5	1	5	2
(mackerel)	liver	Liverpool Bay	8	ND	3	ND
Limanda limanda	flesh	Redear, Yorks	4.6	5.1	9.9	
	flesh	Thames Estuary	2	3	4	0.3
Pleuronectes platessa	flesh	Thames Estuary	3	3	3	0.9
Solea solea	flesh	Thames Estuary	2	4	2	6
(sole)	guts	Thames Estuary	11	1	26	1
Aspitrigla cuculus	flesh	Thames Estuary	11	1	4	0.6
(red gurnard)	guts	Thames Estuary	6	2	10	0.3
Trachurus trachurus (scad)	flesh	Thames Estuary	2	4	1	2
Trisopterus luscus (pout)	flesh	Thames Estuary	2	2	2	0.3
Squalus acanthias (spurdog)	flesh	Thames Estuary	3	1	ND	1

TABLE 5.1. (Continued)

Species		Source	CCl_2CHCl	CCl_2CCl_2	CH_3CCl_3+CCl_4
Scomber scombrus (mackerel)	flesh	Torbay, Devon	2.1	ND	2.4
Clupea sprattus	flesh	Torbay, Devon	3.4	1.0	5.6
Gadus morrhus (cod)	flesh	Torbay, Devon	0.8	<0.1	3.3
	air bladder	Torbay, Devon	<0.1	3.6	NA
		Sea and freshwater birds			
Sula bassana (gannot)	liver	Irish Sea	4.5-6	1.5-3.2	1.2-1.9
	eggs	Irish Sea	9-17	4.5-26	17-20
Phalacrocerax aristotelis (shag)	eggs	Irish Sea	2.4	1.4	39.4-41
Alca torda (razorbill)	eggs	Irish Sea	23-26	19-29	35-43
Rissa tridactyla (kittiwake)	eggs	North Sea	33	25	40
Cygnus olor (swan)	liver	Frodsham Marsh	2.1	1.9	4.7
	kidney	(Merseyside)	14	6.4	2.4
Gallinula chloropus (moorhen)	liver	(Merseyside)	6	3.1	1.6
	muscle	(Merseyside)	2.5	0.7	1.1
	eggs	(Merseyside)	6.2-7.8	1.3-2.5	14.5-21.8
Anas platyrhyncos (mallard)	eggs	(Merseyside)	9.8-16	1.9-4.5	4.2-24

TABLE 5.1. (Continued)

Species		Source	CCl_2CHCl	CCl_2CCl_2	CH_3CCl_3+CCl_4
			Mammals		
Halichoerus grypus (grey seal)	blubber	Farne Is.	2.5-7.2	0.6-19	16-30
	liver	Farne Is.	3-6.2	0-3.2	0.3-4.6
Sorex araneus (common shrew)		Frodsham Marsh	2.6-7.8	1	2.3-7

Source: Pearson and McConnell, 1975.

Note: NA = no analysis; ND = not detectable.

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